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The University of Texas at Dallas
2601 N. Floyd Road
PO Box 830688
Richardson, TX 75083-0688

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14. ABSTRACT

Populations of nuclear spin isomers of 178Hfm2 can be triggered to release their stored energies by the absorption of x-rays having energies in the range 9-13 keV. We have found the basic absorption spectrum for triggering to be approximately 0.16% of the cross section for photoionization of the L-shell electrons surrounding the isomeric nuclei. The maximum effect is induced by X-rays with energies exceeding the threshold for the photoionization of L3 electrons by amounts less than 10 eV. Absorption of a photon of that energy, around 9567 eV triggers the release of 2.446 MeV, thus producing a gain of radiative energy of 256 times for each event. Confidence in the observations of such triggering is better than 10 followed by 1823 zeros to 1.

15. SUBJECT TERMS

Nuclear spin isomers, High energy density materials.

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FINAL PERFORMANCE REPORT

to the

US AIR FORCE OFFICE OF SCIENTIFIC RESEARCH (AFOSR)

describing accomplishments in the

Use of L and M Shell Electrons to Trigger Nuclear Spin Isomers

during the period
March 15, 2003 to September 15, 2007

supported by

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Center for Quantum Electronics
University of Texas at Dallas
PO Box 830688, Richardson, TX 75083-0688

Submitted by the Principal Investigator
Carl B. Collins

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STATEMENT OF OBJECTIVES

Populations of nuclear spin isomers of $^{178}Hf^{m2}$ can be triggered to release their stored energies by the absorption of x-rays having energies in the range 9-13 keV. We have found the basic absorption spectrum for triggering to be approximately 0.16% of the cross section for photoionization of the L-shell electrons surrounding the isomeric nuclei. The maximum effect is induced by X-rays with energies exceeding the threshold for the photoionization of L3 electrons by amounts less than 10 eV. Absorption of a photon of that energy, around 9567 eV triggers the release of 2.446 MeV, thus producing a gain of radiative energy of 256 times for each event.

The period of performance spanned by this report comprised four and one half years of research activity. However, because a manufactured controversy erupted immediately after the commencement of the DARPA SIER Program of which this project was a minor part, a complete redirection of the tasks originally planned for this work became necessary. A politically motivated consortium comprised of past political appointees, Congressional lobbyists, an investigative reporter who engineered the stories about which she wrote and affiliates at Youngstown University succeeded in agitating the Congress to exclude FY 2005 funding for SIER from the Appropriation Bill. They persuasively argued for an impassioned belief that Hf-178 isomers could not trigger. Consequently, the objective of our research was reoriented toward proofs of the triggering of the decay of the $^{178}Hf^{m2}$ isomeric nuclei that could be found with the highest confidence factor possible. That objective had to be pursued without any use of FY05 or subsequent year's money. In this final report here we communicate resulting published results obtained with a confidence of 91σ, an astronomical level of certainty that the $^{178}Hf^{m2}$ nuclear spin isomer has been triggered with soft X-ray irradiation. As can be seen from Appendix 1, that level of certainty corresponds to 1824 "9's" of successful probability.

FINAL STATUS OF HAFNIUM ISOMER TRIGGERING

In most places in the World, irradiation of nuclear isomers of Hafnium-178m2 with soft X-rays will trigger the release of the great amounts of energy stored in the isomer.

Normally, samples of $^{178}Hf^{m2}$ isomeric nuclide have a shelf-halflife of 31-years. They decay by the spontaneous emission of γ photons from a well-known sequence of transitions cascading between the excited states of the nuclei. During such spontaneous decay, the nuclei reach a state with 4 s lifetime about midway through the sequence. However, when triggered by monochromatic X-rays having energies near the L3 edge for photoionization of the surrounding electrons, the induced decay of γ rays follows a different cascade that bypasses the 4 s level and all triggered γ photons are emitted promptly. Different energies of the X-rays cause different sequences of γ rays, but all sequences seem to be emitted promptly. Throughout this work we continued to reexamine the excitation function for prompt decay of the isomer induced by monochromatic synchrotron (SR) X-rays to find energies at which triggering was most efficient using irradiation times at the Japanese SR source SPring-8 that we had won in open competition without cost to this Grant. We found that the maximum effect is induced by X-rays with energies exceeding the threshold for the photoionization of L3 electrons by less than 10 eV. Absorption of a photon of that energy, 9567 eV triggers the release of 2.446 MeV, thus producing a gain of radiative energy of 256X for each event. Measurement of the excitation efficiency around that optimum was achieved with such a number of product photons that statistical confidence reached 32σ for a time-average enhancement of the decay of 5%. Those results have been published in refereed professional journals.

TECHNICAL BACKGROUND

Nuclear spin isomers store the highest densities of energy possible without nuclear reactions. For example, an isomer of ^{178}Hf stores 2.446 MeV per atom for a shelf life of 31 years. [1] In practical terms this means that a sample of the size of a golf ball would store the energy equal to 10 tons of chemical fuels or explosives. However, in nuclear spin isomers the energy is stored electromagnetically so that it would be released as x-rays and γ-rays, when it is triggered. Prior to our work it had been thought to be impossible to trigger the release of these great energy densities trapped in the internal electromagnetic excitations of nuclear spin isomers. ***Building upon our unique level of experience with nuclear spin isomers, during the course of the 5 years of our previous research for AFOSR we demonstrated how to trigger the 31-year isomer of ^{178}Hf.*** [2,3] The x-rays from a small device familiar from dental examinations proved to be sufficient; in fact it is "easy" to trigger nuclear spin isomers. Those extraordinary results sustained peer review, have been published in the leading technical journals [2-5], and have been precisely confirmed by independent work at Sandia National Laboratory. A brief summary of the levels of confidence of both positive and negative results is reproduced in Appendix 1.

Our successes in the use of synchrotron radiation (SR) to trigger nuclear spin isomers have virtually revolutionized the methodology for this type of research. Competing with a prestigious DOE Consortium comprising LLNL, LASL, and the Argonne synchrotron group, as well as the AFOSR-contracted Group at Youngstown University, our Team succeeded at the Japanese facility SPring-8 and at the SLS source of the Paul Scherrer Institute in Switzerland to open completely new directions into isomer triggering. In contrast, the other Consortia produced total failures, because they used SR X-rays of the wrong energy, looked with a detector that was blind to the signature γ photons that are a unique result of triggering, and they looked too late after the irradiation when the γ emission had already finished. It is too early to understand what failed in the Youngstown effort, but it may be instructive to recall that they consistently use a metallized target in which all paths for secondary electrons through the sample are electrically short-circuited. In fact we have continued to perform most of the successful experiments ever conducted on nuclear spin isomers with SR.

In various presentations we have reported that triggering occurs primarily through a mechanism [6,7] mixing properties of the nuclear and atomic states in order to remove the angular momentum retarding the decay of the isomer. In our work the process has been confirmed as a significant effect, being characterized by a cross section approximately equal to 0.16% of the total for the photoionization of the L-shell electrons in Hf. The triggered decay of the ^{178}Hf^{m2} isomer is immediate [8,9]. It bypasses the statistical time lag of the 4 second level that affects all spontaneous decay. The triggering pulses of X-rays from the SR sources last about 20 ps and are separated by about 2 ns. Since fractional "enhancements" of the rate of emission caused by SR X-rays are always measured in terms of time-averages, correcting for the low duty cycle of irradiation means that an enhancement reported as 5% resulted from a 500% increase of the instantaneous rate of gamma emission. These characteristics mean that triggering is very "easy" to accomplish, while nevertheless releasing about 256x the energy expended in triggering. The bandwidth for absorption of trigger photons is very "forgiving," being at least 4 keV wide and the size of the cross section together with the profusion of photons released by each trigger event encourages concepts for applications.

ACCOMPLISHMENTS AND NEW FINDINGS

The landmark results being described in this final report were achieved mostly during the experiments at SPring-8 in June 2004 in a major development of measurement methodology. One of the persistent difficulties with every isomer triggering experiment that uses SR X-rays had been the inability to see anything in real time that correlates with triggering. Were there some immediate diagnostic with which one could detect a signature of triggering, then the experimental parameters could be optimized by maximizing that signature during alignment of the apparatus. During the 2004 experimental series such an immediate diagnostic was proven.

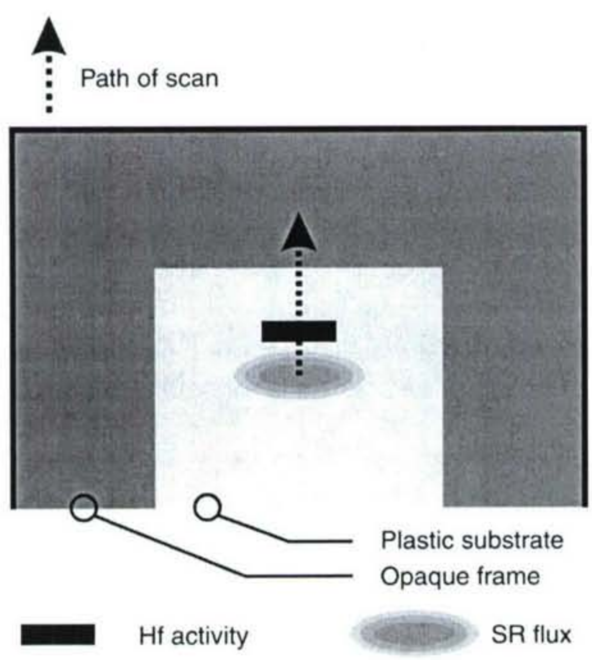

Figure 1 Schematic diagram of the target assembly showing the path that the beam spot traverses during alignment

The alignment of the vertical position of the target with respect to the SR beam of monochromatic X-rays was conducted as shown in Fig. 1. The physical position of the target was moved lower (and farther away) from the Ge detector with a stepping motor so that the relative movement of the "spot" at which the incident beam intercepted the target followed the path shown in Fig. 1 by the arrow. Fig. 2 shows the result of measurements of the total numbers of γ-photons detected each 300 s that were also displayed in real time on a monitor as the physical position was changed. The increase of the rate at which γ-photons were detected clearly shows where the incident spot passed across the isomeric content of the target. Measurement at a single position for 300 s showed that a 5% increase of emission was induced by the incident X-rays. For that alignment the flux was 2 x 10^{12} cm^{-2} s^{-1} at 9567 eV. Spectral linewidth of the X-rays was 1 eV. The actual numbers of photons are shown in Table 1. The confidence that incidence of the monochromatic X-rays upon the isomeric content of the target caused an enhanced collection of γ-photons is 91σ, a strong

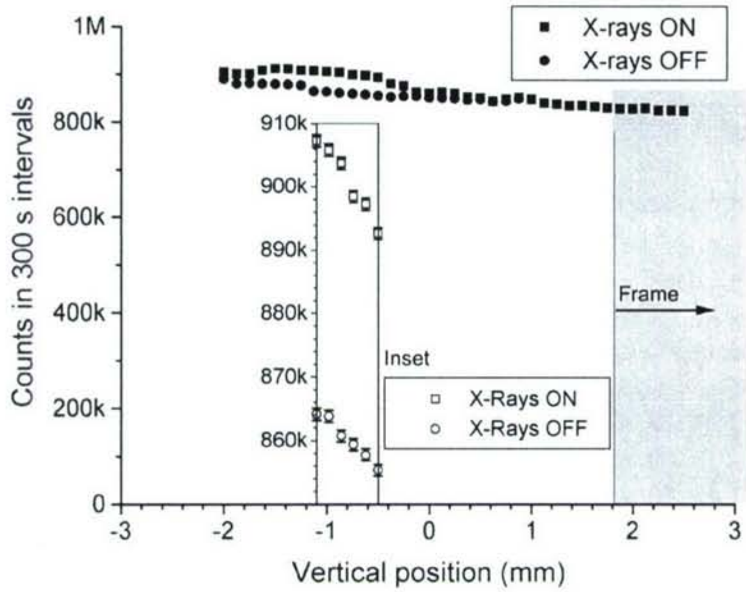

Figure 2 Graph of the numbers of γ-photons counted during each 300 s during which the target remained in the position shown on the abscissa. Inset is magnified to show the error bars

confirmation of our triggering of the decay. The confidence of the maximum enhancement being 5% is seen to be 32 σ. This datum is shown as the red triangle in the summary of confidences shown in Appendix 1. Also, it can be seen that nothing perceptible was found when the irradiation passed over the substrate, nor when it intercepted the relatively massive brass frame of the target.

Now it is possible to optimize the apparatus and methodology for isomer triggering with high confidence by using displayed data that is visible in real time.

Incidence Position	Activity max. -0.9 mm	Activity -2.0 to -0.2 mm	Substrate 0.6 to 1.0 mm
Beam On	903709 (951)	13505081 (3675)	2542876 (1595)
Beam Off	860729 (928)	13033053 (3610)	2536650 (1593)
Increase	42980 (1328)	472028 (5152)	6626 (2254)
Increase	5.0 (0.15)%	3.6 (0.04)%	0.26 (0.09)%
Sigma	32.4 σ	91.6 σ	2.9 σ

Table 1 Tabulated results of the measurement of total gammas counts collected in 300 s intervals for different positions of the target relative to the beam spot of SR monochromatic X-rays with energy 9567 eV and with an average flux of 2×10^{12} cm^{-2}s^{-1}. Notable are the results from summing enhanced numbers of counts over the relative positions ranging from -2.0 to -0.2. Average enhancement is 3.6% with a confidence of 91.6 σ. The maximum enhancement of 5% with a confidence of 32.4 σ is found in the leftmost column corresponding to the position -0.9 mm. Vertical positions are keyed to dimensions in Fig.2

Analysis of the Japanese Spring-8 data also showed several new lines in the γ ray emission spectrum from triggered Hf-isomers. In most cases these new lines could not be attributed to particular transitions between known excited states of the isomer. However, one new line at 2457.2 keV was more energetic than the 2446.05 keV energy stored by the isomer. That new line must have resulted from a transition from a level lying above the isomer in energy. Details are found in the Ref. [10], a copy of which appears in Appendix 2.

The SR sources radiate considerable electromagnetic interference (EMI) and their use in nuclear fluorescence measurements is still far from common. Because of the continued appearance of unexpected results from the irradiation of isomeric samples, it was decided to develop a digital simulator for use in calibrating any future experiments. Such a project was feasible without the use of any FY05 monies.

"Null" experiments had already been done, but without a radioactive target there was not much signal to examine for EMI that might distort a measurement of γ counting rates. To improve upon a search for unexpected aberrations from EMI a simulator was developed that produced successive random bursts of time-dependent voltages that precisely mimicked the output voltages from an Ortec 673 spectroscopy amplifier. It was capable of generating the data stream corresponding to the emission of random pulses the amplitudes of which corresponded to one of 10 possible transitions being simulated. Both energies and relative strengths of the 10 selected transitions could be selected by the operator. The intent was to develop and document a device that could be used to generate a precisely known sequence of pulses of the type obtained from nuclear spectroscopy amplifiers used to record the γ emission from a radioactive sample. It was to be used in later experiments to determine the degree of modulation of the simulated outputs when a proximate SR source was being used. Collection of simulated data showing no change when the SR beam was "ON" would further confirm confidence in the particular measurement technology being used in these isomer triggering experiments.

For illustrative data shown here, the average random photon rates were set to 3 kcps and the spectral complexity was chosen to simulate 5 lines being independently emitted with relative intensities covering one order of magnitude.

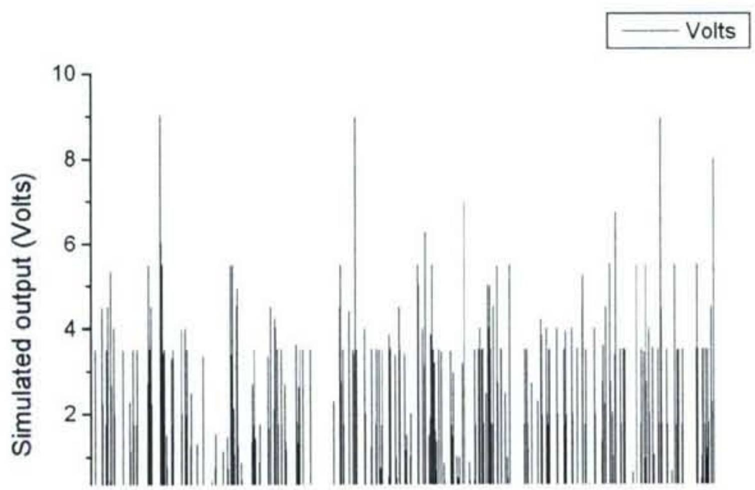

Figure 3 Simulator output voltage.

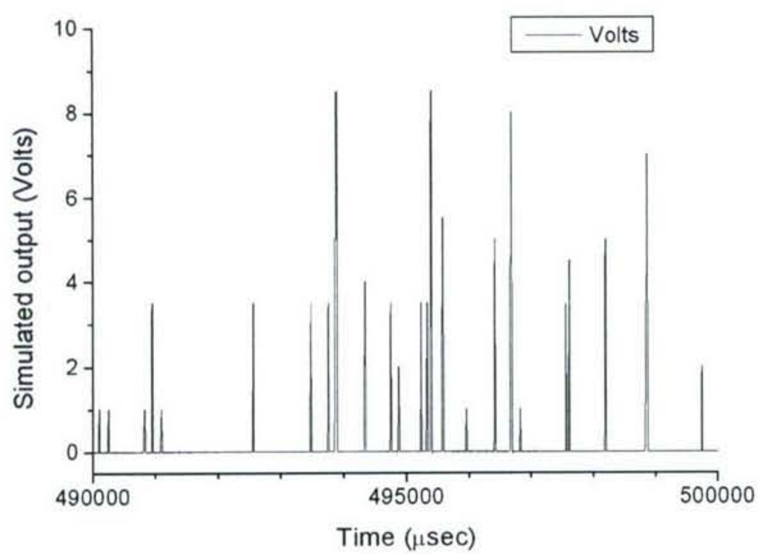

Figure 4 Data of Fig. 3 plotted on a magnified abscissa.

Figure 3 shows part of the "raw" output from the simulator. Since the sampling rate was 700 kHz, not all discrete "points" of output voltage are seen in the plot Successive magnifications of the data of Fig. 3 follow. They show the distributions and behaviors as regards "pile-ups" of the sorts that are seen in actual data..

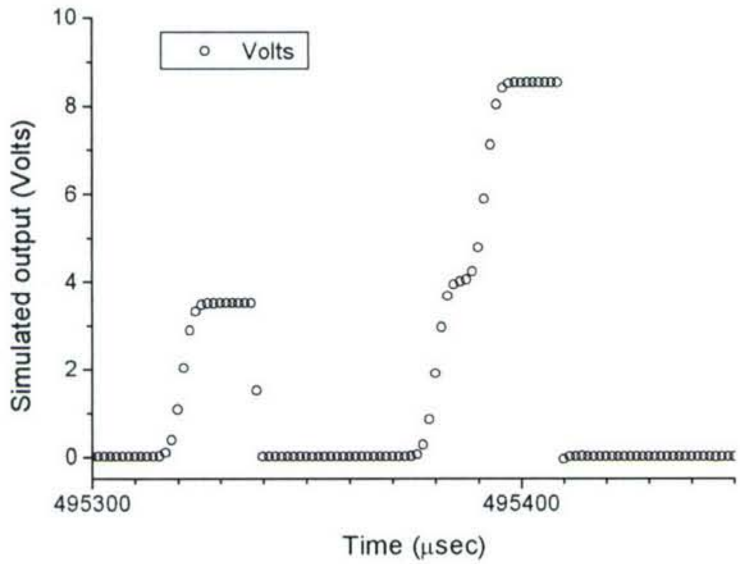

Figure 5 Further magnification showing the size of the mesh over which the simulated output pulses are generated.

The conclusion of this facet of the work is that the digital simulator is ready for use in further building confidence that the data logging method used to record the raw output voltages from the spectroscopy amplifiers as functions of experimental times is recording all of the data and adding nothing. No anomalies were found in the data acquisition system.

The overall conclusion of this work is that the confidence that the Hf-isomer is triggered by incident X-rays has been demonstrated at a level of 91σ. As can be seen from Appendix 1, that corresponds to 1824 "9's" of probability.

BUT DOESN'T THIS MEAN THAT THE LAWS OF PHYSICS ARE VIOLATED ?

NO! One of the facile, but popular, criticisms of these results is that they are somehow, "too good to be true" and so must violate basic laws of Physics. When an X-ray photon is absorbed by a heavy element such as Hafnium sometimes an electron is "knocked out" of the atom leaving an L-shell vacancy. In about 23% of the cases of L3 shell holes, another electron "falls" into the vacancy and an X-ray is emitted. Most of the rest of the time the vacancy is filled by rearrangement of other electrons some being ejected from the atom and thus carrying away the energy of the vacancy without the emission of an X-ray. As we reported in several peer-reviewed articles, the fraction of the cases of L3 shell photoionization that end in a nuclear event that triggers a release of the stored energy of the isomer is only 0.16%; only 16 times in 10,000 events. It's a trivial addition to the well-known channels for the absorption of X-rays.

A more sophisticated assertion that the laws of physics must forbid isomer triggering is founded in the absolute faith in the conservation of the quantum number called K attributed to various states of Hafnium nuclei. The Hafnium nucleus is not a sphere but more of a football shape and that number K is the degree to which the rotations of the protons and neutrons in the nucleus are supposed to go around the long axis of the "football." For ideal nuclei, big changes of K during a process are supposed to hinder it by large factors. So theoreticians believe the laws of physics demand "K-hindering" of all processes that would require a large change of K, in ideal nuclei and in perfectly spherical elephants. However, in Russia, K-hindering has just been measured to be weak to non-existent in the nuclear spin isomer of Hf-178m2. There is no K-hindering in the Hafnium-178m2 isomer. Those results have been published in one of the elite, peer-reviewed US journals, [11] but have been completely ignored by those who prefer to believe in more idealized models of nuclei.

In summary, the rate of Hafnium isomer triggering is only an unexpected 16 parts in 10,000 adding to the atomic physics of X-ray absorption. It is completely allowed because Hafnium is not much like the "ideal" model nucleus for which K-hindering would have been violated. As reviewed in Appendix 1, there have been many more high-confidence reports of hafnium isomer triggering than there have been failed experiments to demonstrate it.

The nuclear isomer of Hafnium-178m2 triggers and triggers rather easily, and no laws of physics are violated when it triggers.

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PERSONNEL SUPPORTED

Professional personnel supported and/ or associated with this project during the current reporting period have been:

1) Carl B. Collins	UTD Professor and Principal Investigator
2) Farzin Davanloo	UTD Research Scientist
3) C. Zoita	UTD Graduate Student
4) J. M. Pouvesle	Research Director, GREMI, Univ. d' Orleans, France

PUBLICATIONS

Reviewed publications that include presentations of the accomplishments for AFOSR during this reporting period. The last is reproduced in Appendix 2 for convenience.

"Accelerated γ-emission from isomeric nuclei" by C. B. Collins, N. C. Zoita, F. Davanloo, S. Emura, Y. Yoda, T. Uruga, B. Patterson, B. Schmitt, J. M. Pouvesle, I. I. Popescu, V. I. Kirischuk, and N. V. Strilchuk, Radiat. Phys. and Chem. **71**, 619 (2004).

"Nuclear resonance spectroscopy of the 31-year isomer of Hf-178" by C. B. Collins, N. C. Zoita, F. Davanloo, Y. Yoda, T. Uruga, J. M. Pouvesle, and I. I. Popescu, Laser Phys. Lett. **2**, 162 (2005).

INTERACTIONS/TRANSITIONS

For this reporting period, interactions have been intense, but informal. The most notable level has been sustained with colleagues at Sandia, in particular Dr. Pat McDaniel with whom frequent consultations continue.

HONORS / AWARDS

The Principal Investigator, Prof. C. B. Collins has received the following lifetime honors and awards:

1960	Woodrow Wilson Fellowship
1972	Fellow of the American Physical Society
1984	Senior Member of the IEEE
1989	1989 Distinguished Texas Scientist Award from the Texas Academy of Sciences
1992	Doctor Honoris Causa, University of Orleans, France
1993	Romanian Academy of Sciences, Honorary Member

Appendix 1

Reprint of the Letter
being circulated upon request

Summarizing the confidence of all Hf-isomer triggering experiments

by C. B. Collins

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Appendix 2

Reprint of the Publication

"Nuclear resonance spectroscopy of the 31-year isomer of Hf-178"
by
C. B. Collins, N. C. Zoita, F. Davanloo, Y. Yoda, T. Uruga, J. M. Pouvesle, and I. I. Popescu

Laser Phys. Lett. **2**, 162 (2005).

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